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PATENT SPECIFICATION

954,594

DRAWINGS ATTACHED.

Inventor:—JAN VITESLAV WEINBERGER.



954,594

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June 12, 1962.

No. 22496/62.

Complete Specification Published : April 8, 1964.

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Index at Acceptance :—G6 R2.

International Classification :—G 21.

COMPLETE SPECIFICATION.

Flexible Shield for Ionizing Radiations.

We, GENTEX CORPORATION, a Corporation duly organized and existing under the laws of the State of Delaware, United States of America, and having a principal place of business at 450 Seventh Avenue, New York, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:

This invention relates to a flexible shield for ionizing radiations and, more particularly, to an improved flexible medium for providing protection from the emanations of radioactive substances, X-rays and the like. Owing to the fact that ionizing radiations are becoming more and more prevalent in industry as well as in medicine and owing to the fact that such emanations may do serious injury to human beings if subjected to over-doses of the same, it is becoming increasingly important to find materials which will shield against ionizing radiations, which materials may be flexible so that they may be shaped to parts of the body and used for gloves, helments, aprons, leggings, clothing and the like.

Such ionizing radiations vary depending on the energy level of the radiation and include alpha particles, protons, neutrons, positrons, beta rays and gamma rays.

Lead, owing to its high molecular weight, is an excellent shielding material. Unfortunately, however, where a shield has to be contoured to fit a patient's body as in cases where it is desired to subject a particular area of the body to ionizing radiations it is difficult to form a sheet of lead to the required shape and it is infeasible to have a number of shields for different

sizes of persons for whom shields are required. It is also extremely desirable to shield the bodies of persons who are occupationally exposed to radiation as, for example, X-ray technicians, medical men employed around X-ray machines, maintenance crews for reactors and technicians who deal with radioactive isotopes.

In our copending Application No. 1499/62 (Serial No. 954,593), there is disclosed a flexible shield for ionizing radiations which employs a flexible carrier provided with a coating of an amalgam of lead and mercury.

While such shield is effective mercury is expensive and the flexibility is of limited degree.

We have now discovered it is possible to form a shield for ionizing radiations by means of an improved material which is inherently flexible and moldable in and of itself. It may be secured to a fabric or sandwiched between layers of fabric to provide a flexible material which may be readily fashioned into clothing and flexible shields for ionizing radiations.

According to the present invention we provide a flexible material for absorbing ionizing radiation which comprises a major proportion by weight of powdered lead, a minor amount by weight of silicone rubber and a lesser amount by weight of fiber flock. Further, according to the invention we provide a flexible shield for ionizing radiations which comprises a major proportion by weight of powdered lead, a minor amount by weight of silicone rubber and a lesser amount by weight of fiber flock.

Preferred embodiments of the invention are hereinafter particularly described, by

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way of example only, with reference to the accompanying drawings, in which:—

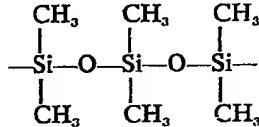
Figure 1 is a sectional view of an assembly showing one embodiment of the invention.

Figure 2 is a sectional view of a multi-laminate assembly showing another form of the invention.

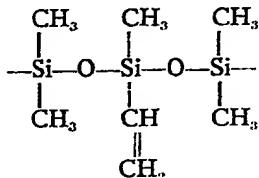
Advantageously, the lead powder is such that the preponderance of it will pass through a number 200 Tyler mesh sieve. Such powder may have a minimum lead content in excess of 99.9% by weight of lead together with traces of silver, copper, arsenic, antimony, tin, zinc, iron and bismuth. Lead powder is well known to the art and is produced by atomization.

Most shielding materials of the prior art are bulky owing to the fact that the art has been unable to exceed a lead content in the vicinity of 40% by weight with relation to the carrier. In one embodiment of the invention a lead contact as high as 98.5% by weight may be employed owing to the use of only 1% by weight of silicone rubber and $\frac{1}{2}$ of 1% by weight of fiber flock. The large quantity of lead per weight with respect to the small quantity of binder produces a high degree of shielding and yet the material is sufficiently flexible to be adhered to or otherwise secured to fabric to enable the production of flexible clothing.

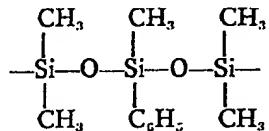
The silicones are semi-inorganic polymers and produce dense, rubber-like material. The most common type of silicone rubber is polydimethylsiloxane. It has the general structure shown by the following fragment of the chain.



In a second type of silicone rubber a small percentage of the methyl groups have been replaced by vinyl groups. A fragment of the chain has the following appearance.



In a third type of silicone rubber a small percentage of phenyl groups are substituted for the methyl groups. A fragment showing the chain is as follows.



It is understood, of course, that the molecular weight of silicone rubber ranges from approximately 300,000 to about 1,000,000.

Any of these silicone rubbers may be employed in carrying out the invention but the use of a vinyl type improves vulcanization properties.

For flock, any fibrous material such as wool, cotton, rayon or any of the synthetic extruded fibers such as polyethylene or nylon may be used. Advantageously, fiber flock produced from wool can be used. Fibers of coarse wool easily separate from each other and wool has the highest natural moisture regain being in the order of 15% to 18% as compared with 8% for cotton and $\frac{1}{2}$ of 1% to 4% for fibers of the thermoplastic family. The hydroscopic characteristic of wool facilitates the curing of silicone rubbers.

We have also found that conventional rubber cannot be readily amalgamated with lead to form a homogeneous compound. It is only by the use of silicon rubbers that we have been able to achieve a homogeneous amalgamation of the lead and the rubber. Apparently silicon rubber has the property of removing the lead oxide layer on the lead powder and enables a remarkably homogeneous compound to be made.

In carrying out the invention a dough was formed by mixing lead powder described above in the amount of 98.5% by weight with 1% by weight of silicone rubber of type 2 above and $\frac{1}{2}$ of 1% of fiber flock comprising largely wool fibers. The mixture was deposited on a sheet of vinylidene chloride copolymer. It was then thinned by adding toluene in an amount of 5% by weight of the mixture. The mixture was then molded with a blade coated with tetrafluoroethylene copolymer, e.g. "Teflon" (Registered Trade Mark). This was necessary owing to the fact that the material adheres very strongly to metals. In making large quantities of the material mixers lined with polytetrafluoroethylene must be employed.

During the mixing the curing agent known to the art for silicon rubber, namely benzoyl peroxide, is added. After the material has been thoroughly mixed a sheet is formed by rolling the material with a 105 rolling pin covered with polytetrafluoroethylene in a manner of rolling out a sheet of dough in baking, the rolling, of course, being carried out against the vinylidene chloride copolymer sheet. The rolling is 110

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continued to produce a sheet of the dough having a thickness of about $1\frac{1}{2}$ millimeters. In making large quantities of the material the sheets would be rolled by calender rolls coated with polytetrafluoroethylene. The interroll distance will determine the thickness of the sheet of mixture.

The curing may be carried out in two stages. In the first stage the cure is advanced to a point at which the rolled sheet has dimensional stability under its own weight. This can be readily accomplished by subjecting the sheet to a temperature of about 260° F. for a period of between 5 to 10 minutes. It is understood, of course, that the sheets can be contoured to any desired shape by molds and cured in the molds for 5 or 10 minutes to give the piece dimensional stability. The molded piece is then transferred to a well ventilated oven and baked several hours at about 480° F. This dries off the breakdown fragments from the peroxide and the light gum fractions. The cure also establishes 25 additional cross linkages.

It is understood, of course, that if desired room temperature curing silicones can be used. These are achieved by adding accelerators such as dithiocarbamates, carbon disulfide or zinc mercaptobenzothiazole.

In another embodiment of the invention the above procedure was followed with the exception that xenol was used as a solvent. Most of the solvent it will be found will evaporate during the mixing of the dough. The basic material thus formed is then assembled with the fabric. It may be adhered to a fabric by any suitable material. We have found that advantageously 90% by weight of silicon rubber may be mixed with 10% by weight of fiber flock such as wool or cotton, preferably using a silicon rubber material of the room temperature curing type. This material is known to the art and contains the necessary catalysts and accelerators for curing at room temperature. The silicone rubber is shipped and stored in air tight containers and usually contains moderators to prevent vulcanization in storage as is well-known in the art. The flock is mixed with this silicone rubber and the resulting mixture forms the adhesive by which a textile layer may be adhered to the basic cured sheet of lead silicone rubber.

Referring now to Figure 1, the layer 10 comprises sheet containing 98.5% by weight of lead, 0.5% by weight of fiber flock and 1.0% by weight of silicone rubber. It is formed and cured as pointed out above. The layer 10 which contains the lead and forms the barrier for stopping the ionizing radiations is protected by 65 an upper layer of fabric 12 and a lower

layer of fabric 14. These layers of fabric are adhered to the barrier layer 10 by layers of adhesive 16 and 18. It is understood, of course, that any appropriate adhesive can be used such as rubber cement or neoprene cement. Advantageously, a mixture comprising 90% by weight of silicon rubber and 10% by weight of fiber flock is employed. This silicone rubber and fiber flock not only secures the fabric to the barrier layer 10 but also provides better tensile strength and resistance against cracking and stress.

The adhesive layers 16 and 18 can be cured by heating or can be of the room temperature curing type described above.

The fabric layers 12 and 14 may be of any appropriate fabric. Advantageously the heat resistant cloth shown in U.S. Patent No. 2,884,018 is employed for this purpose.

If the fabric is made of the heat resistant type it will be apparent that the flexible shielding material can be made into garments or molded into shapes conforming to curvature of the body and thus provide protective clothing guarding against nuclear flash resulting from atomic explosions.

If one of the fabric layers 12 or 14 be made of the heat resistant cloth shown in the above U.S. Patent the adhesive layers 16 and 18 must be of the room temperature curing type owing to the fact that the curing temperature may deleteriously affect the material of which the heat resistant fabric is formed. Stated otherwise, a component of the heat resistant fabric sublimates at elevated temperatures to enable the heat resistant mechanism to occur. It is understood, of course, that this mechanism must not be permitted to occur by the elevated temperature of curing.

Referring now to Figure 2, it will be seen that two barrier layers 10 are provided each of which is formed of the lead powder cohered by silicone rubber. The upper layer 12 of the fabric is similar to the layer 12 in Figure 1 and the lower layer of fabric 14 is similar to the layer 12 in Figure 1. It is understood, of course, that one of the layers 12 or 14 or both may, if desired, be made of heat resistant fabric as pointed out above. The adjacent barrier layers 10 are adhered by an intermediate adhesive layer 20 which advantageously may be made of 90% by weight silicone rubber and 10% of fiber flock. The adhesive layer 16 and the adhesive layer 18 in Figure 2 are similar to the corresponding adhesive layers 16 and 18 of Figure 1. The barrier layers 10, as pointed out above, may advantageously be in the vicinity of between 1 millimeter and $1\frac{1}{2}$ millimeters in thickness though it is to be 130

understood, of course, that any desired thickness may be employed. We have found that the employment of a multiplicity of thinner layers makes a more flexible though more bulky material.

If a permanently curved form is desired a single thicker layer will be employed and molded to the desired form and cured in the molded shape.

The finished material shown in both Figures 1 and 2 may be calendered to uniform thickness and then formed into the desired protective clothing.

It might be thought that the use of finely divided lead particles such as powdered lead bonded into a homogeneous material by silicone rubber might not be efficacious in stopping ionizing radiations owing to the interstices between the lead particles.

In order to determine whether or not the construction which imparted flexibility sacrificed shielding power as shown in the embodiment of Figure 1 was made in which the barrier layer 10

had a thickness of 1.5 millimeters. The density per unit area of the material was 1.25 grams per square centimeter. A "Kodak" (Registered Trade Mark) industrial X-ray film type KK was subjected to

100 kv unfiltered X-rays under broad beam conditions for 10 seconds exposure at 0.64 r/seconds which X-ray film was shielded with the sample of the material. Under these conditions we found that there

was transmitted through the material only $0.22\% \pm 0.01\%$ of the X-rays. The lead equivalence was determined by replacing the sample of material in the X-ray beam with various thicknesses of lead; the lead equivalence was 1.06 millimeters of lead $\pm 0.02\%$.

The same test was conducted with 250 KV unfiltered X-rays and obtained an X-ray transmission of $4.1\% \pm 0.2\%$ through the sample. This was equivalent to 1.11 millimeters of lead $\pm 0.02\%$.

The results of the test prove that exceptionally high X-ray absorption is achieved and that the fine subdivision of the lead particles together with the minor amount of silicone rubber which need be employed permits a sufficiently close packing so that excellent X-ray absorption is obtained while achieving the desired flexibility.

The material may be readily molded into

any desired shape so that it may be employed to shield the bodies of persons who are occupationally exposed to radiation and to protect curved parts of the body when it is desired to subject certain other areas to ionizing radiations in connection with medical therapy.

The material can be made abrasive resistant and readily combined with heat resistant fabric.

WHAT WE CLAIM IS:—

1. A flexible material for absorbing ionizing radiation which comprises a major proportion by weight of powdered lead, a minor amount by weight of silicone rubber and a lesser amount by weight of fiber flock. 70

2. A material according to Claim 1 comprising 98.5% by weight lead, 1.0% by weight silicone rubber and 0.5% by weight fiber flock. 75

3. A flexible shield for ionizing radiations which comprises a major proportion by weight of powdered lead, a minor amount by weight of silicone rubber and a lesser amount by weight of fiber flock. 80

4. A shield according to Claim 3 comprising 98.5% by weight lead, 1.0% by weight silicone rubber and 0.5% by weight fiber flock. 85

5. A shield according to Claim 3 or 4 in which the lead, rubber and flock are adhered to a layer of fabric. 90

6. A shield according to Claim 5 in which the lead, rubber and flock are sandwiched between two layers of fabric by means of an adhesive. 95

7. A shield according to Claim 5 or 6 in which the adhesive comprises 90% by weight silicone rubber and 10% by weight fiber flock. 95

8. A shield according to any of Claims 5 to 7 in which at least one of the layers of fabric is a heat resistant cloth. 100

9. A flexible shield for ionizing radiations substantially as herein described with reference to the accompanying drawings.

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London, W.C.1,
For the Applicants.

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COMPLETE SPECIFICATION

1 SHEET

*This drawing is a reproduction of
the Original on a reduced scale*

Fig 1

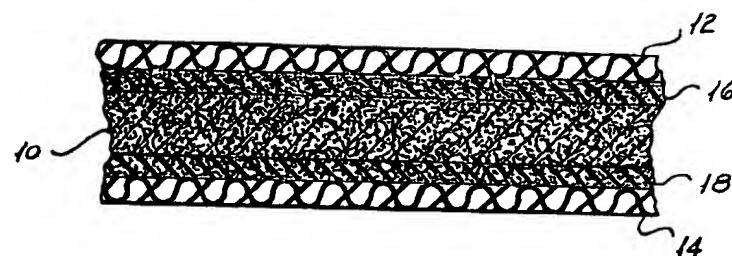


Fig 2

